

## Supporting information

for

### **Robust bifunctionality towards hydrogen evolution and ethanol oxidation reactions catalyzed by six-element high entropy alloys**

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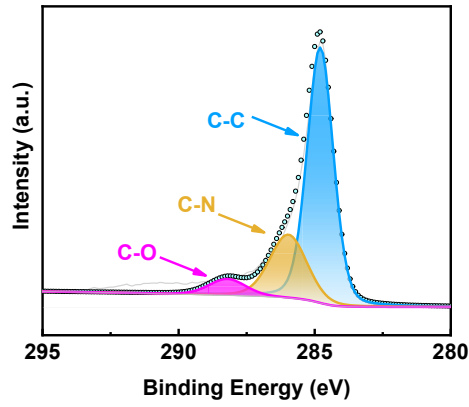
## **Experimental section**

**Synthesis of FeCoNiIrPtPd/NCNT:** A solution was prepared by mixing dicyandiamide (1.5 g),  $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$  (0.2 mmol),  $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$  (0.2 mmol),  $\text{NiCl}_2 \cdot 6\text{H}_2\text{O}$  (0.2 mmol),  $\text{PdCl}_3$  (0.1 mmol),  $\text{IrCl}_3$  (0.1 mmol),  $\text{H}_2\text{PtCl}_6$  (0.1 mmol) and 80 mL of ethanol, and the mixture was stirred at room temperature (25 °C) for 2 h. The mixture was then heated to 80 °C and stirred continuously until the solvent evaporated. The resulting solid was ground into a fine powder using a mortar and pestle, then placed in a tube furnace. It was heated under a nitrogen atmosphere at a rate of 10 °C per minute up to 800 °C, where it was maintained for 2 h, producing the FeCoNiIrPtPd/NCNT electrocatalyst. The NCNT was produced through the treatment of Fe/NCNT with a strong acid.

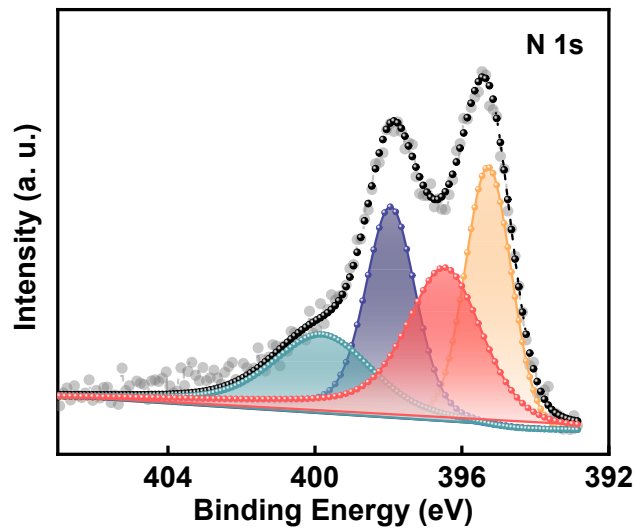
**Fundamental Characterizations:** The phase of the samples was analyzed using X-ray diffraction (XRD) patterns from a Bruker D8 Advance instrument. The structure of the electrocatalyst was characterized using X-ray photoelectron spectroscopy (XPS) (Thermo Fisher, USA). The microscopic morphology of the samples was observed with a field emission scanning electron microscope (SEM, SU8010, Japan), and the microstructure and elemental distribution were analyzed using high-resolution transmission electron microscopy (TEM, FEI Themis 300, USA).

**Ink Preparation and Electrochemical Evaluation:** The ink was prepared by ultrasonically dispersing a mixture of the synthesized catalyst (2 mg) and carbon black (2 mg) in 780  $\mu\text{L}$  of deionized water, 200  $\mu\text{L}$  of isopropanol, and 20  $\mu\text{L}$  of 5 wt% Nafion solution for over 30 minutes. A 10  $\mu\text{L}$  aliquot of this ink was evenly applied

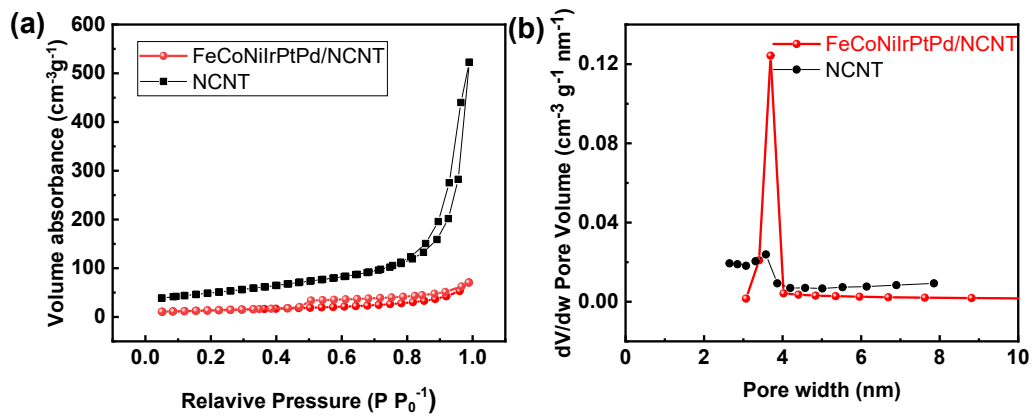
onto the surface of a glassy carbon electrode (GCE, 3 mm diameter) to form the working electrode. Electrochemical tests were then conducted at room temperature (25 °C) using a Gamry potentiostat (Interface 1000E, USA) with a standard three-electrode system, which included a carbon rod as the counter electrode and a Hg/HgCl<sub>2</sub> electrode as the reference electrode. The scan rate was 5 mV s<sup>-1</sup> for recording LSV curves.



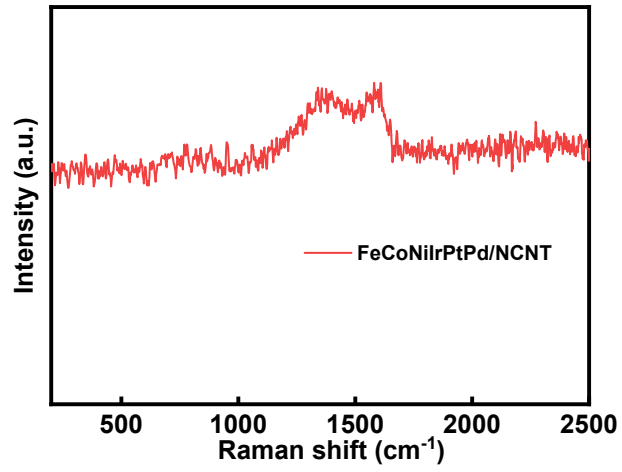
**Figure S1** C 1s XPS core level spectrum of FeCoNiIrPtPd/NCNT.



**Figure S2** N 1s XPS spectrum of FeCoNiIrPtPd/NCNT.



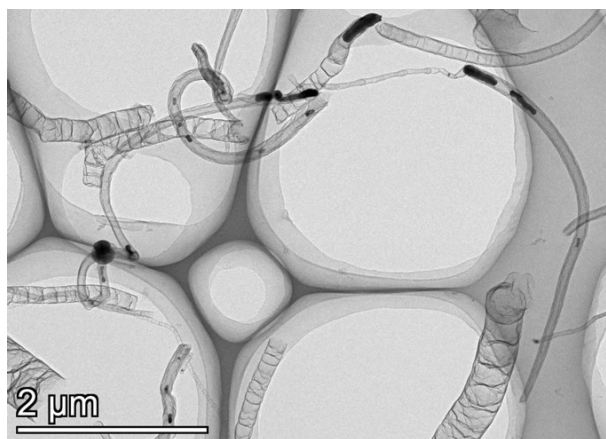
**Figure S3** N<sub>2</sub> isothermal curves (a) and pore size distribution (b) of FeCoNiIrPtPd/NCNT and NCNT.



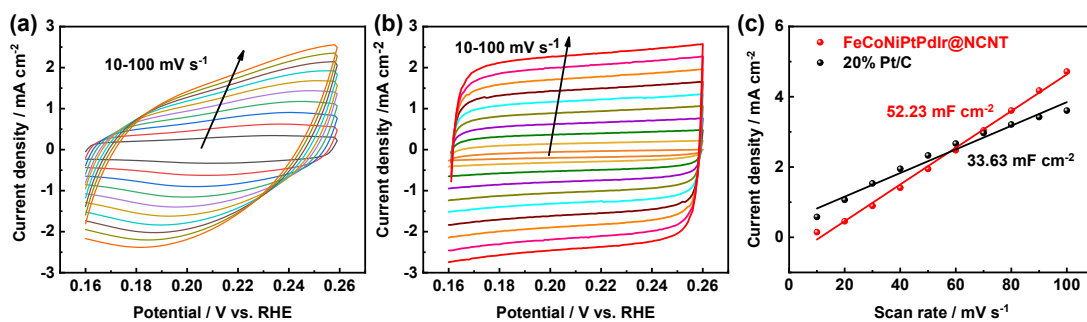
**Figure S4** Raman spectroscopy of FeCoNiIrPtPd/NCNT.



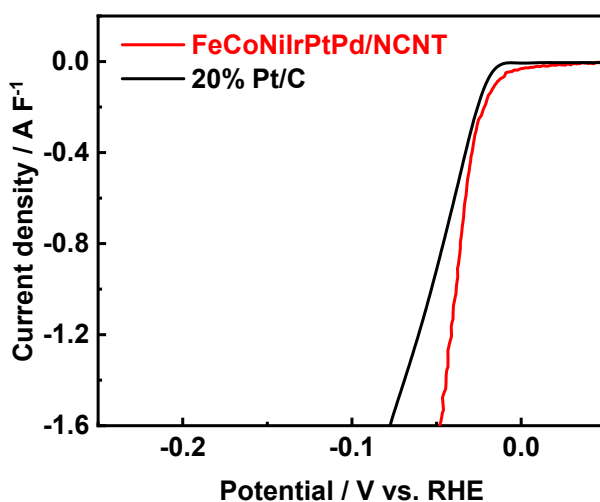
**Figure S5** SEM image of NCNT.



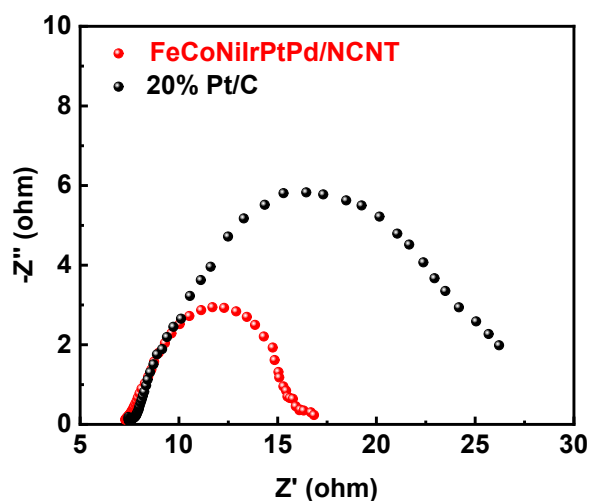
**Figure S6** TEM image of FeCoNiIrPtPd/NCNT with low magnification.



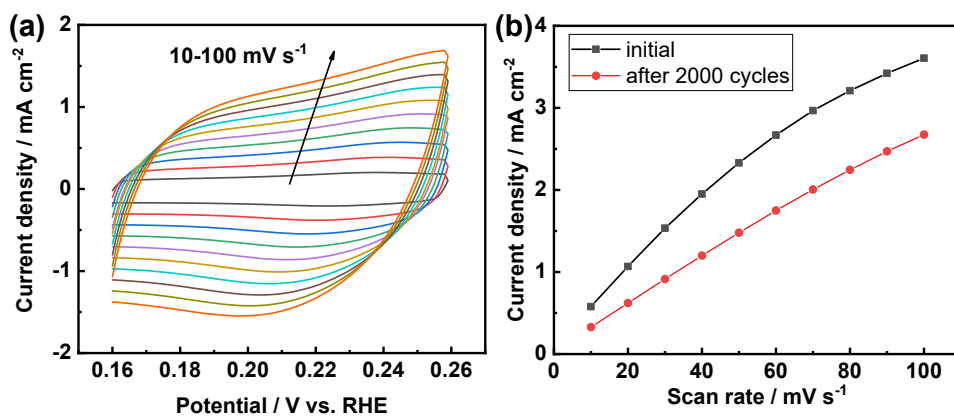
**Figure S7** Cyclic voltammetry curves of Pt/C (a) and FeCoNiIrPtPd/NCNT. (c)  $C_{dl}$  of Pt/C and FeCoNiIrPtPd/NCNT.



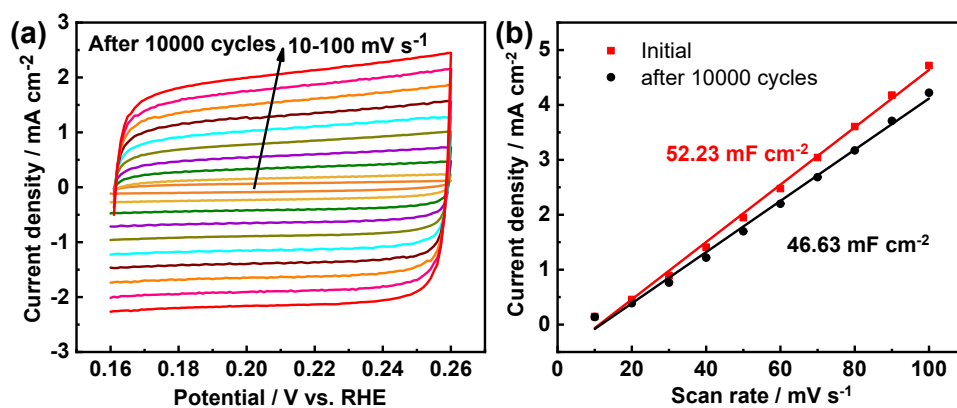
**Figure S8** Specific activity of FeCoNiIrPtPd/NCNT and Pt/C in HER catalysis.



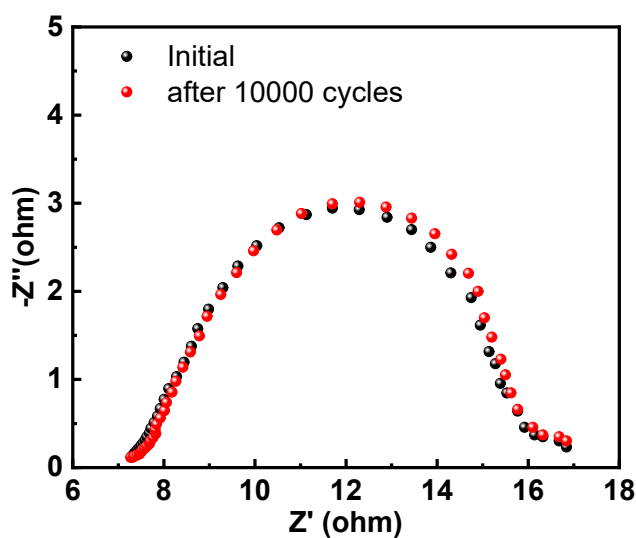
**Figure S9** Electrochemical impedance spectroscopy of FeCoNiIrPtPd/NCNT and Pt/C.



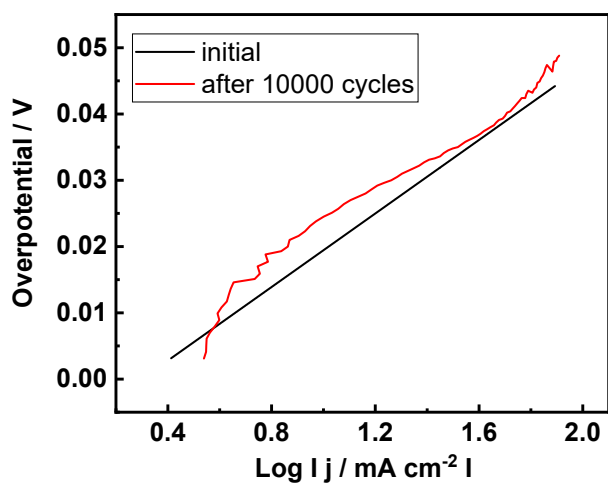
**Figure S10** Cyclic voltammety curve of Pt/C after 2000 cycles. (b)  $C_{dl}$  of Pt/C before and after 2000 cycles.



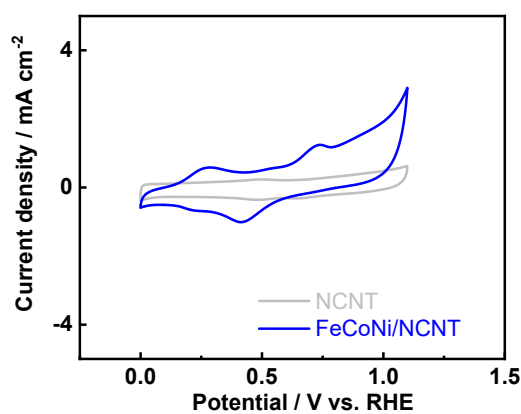
**Figure S11** Cyclic voltammety curve of FeCoNiIrPtPd/NCNT after 10000 cycles. (b)  $C_{dl}$  of Pt/C before and after 2000 cycles.



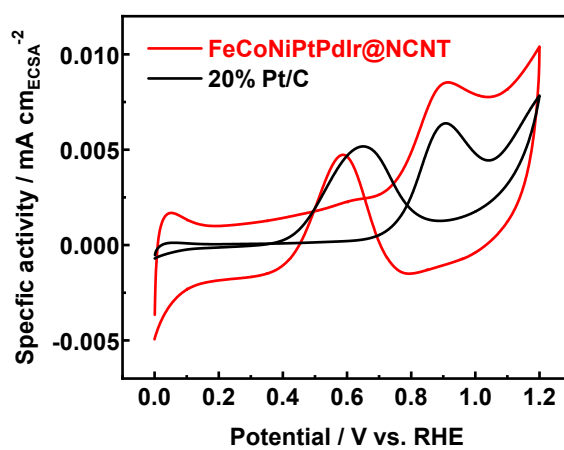
**Figure S12** Electrochemical impedance spectroscopy of FeCoNiIrPtPd/NCNT before and after 10000 cycles.



**Figure S13** Tafel slope of FeCoNiIrPtPd/NCNT before and after 10000 cycles.

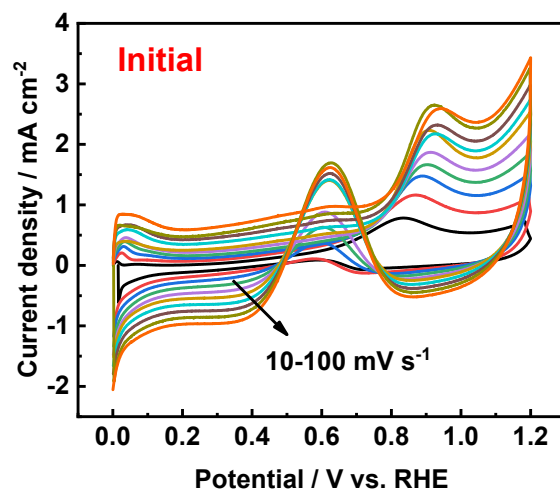


**Figure S14** EOR performance of NCNT and FeCoNi/NCNT.

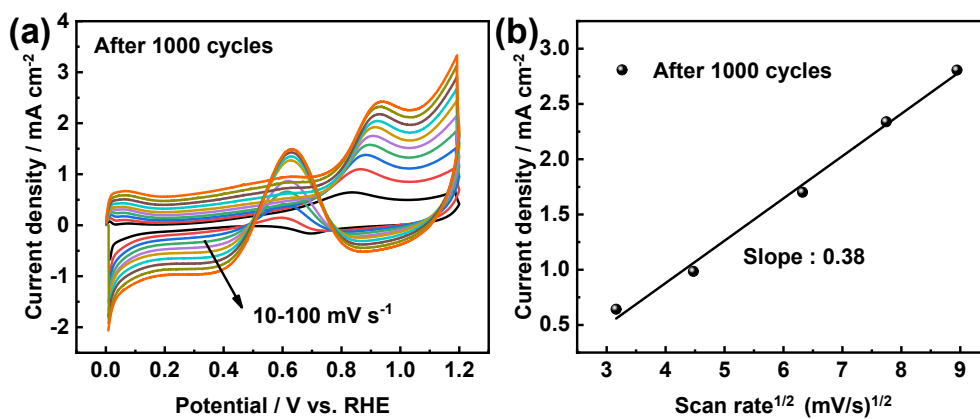


**Figure S15** Specific activity of FeCoNiIrPtPd/NCNT and Pt/C in EOR catalysis.





**Figure S16** EOR performance recorded at different scan rates of FeCoNiIrPtPd/NCNT.



**Figure S17** EOR performance recorded at different scan rates after 1000 cycles. (b) Slope of peak current density vs. scan rate for FeCoNiIrPtPd/NCNT after 1000 cycles.